Growth Rates of New Grains during the Primary Recrystallization of Aluminium Plates

Growth Rates of New Grains during the Primary Recrystallization of Aluminium Plates*

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Synopsis

The growth rates of recrystallized grains in cold worked aluminium plates were measured by means of the method employed by Anderson and Mehl with slight modification. The measurements were carried out under various circumstances, especially investigating the effects of impurity, temperature, deformation and grain size on the growth rates over extensively wide range of these variables. The measurements offer important informations concerning these effects, which will be able to improve the atomistic theory of grain boundary migration.

I. Introduction

In recent years our knowledge concerning the primary recrystallization has considerably been improved by the studies on this subject in terms of the rate of nucleation $\dot{N}$ and of the rate of growth G. The most satisfactory measurements on these two basic rates were carried out by Anderson and Mehl(1). They investigated the effects of temperature, deformation and grain size on both kinds of rate by using high purity aluminium. In Anderson and Mehl's procedure, G was determined by plotting the radii of the largest grains (which had not impinged on another growing grain) observed in a series of samples recrystallized for different time periods, under the assumption that the largest grains nucleated at the same time instant.

Similar measurement had been carried out by Kornfeld and Pawlow on aluminium wires at 1937(2). They, however, measured at temperatures higher than 400°C, while Anderson and Mehl measured at the lower than 400°C. There are considerable divergences between the results of these two investigations.

Fujiwara and his co-workers(3) measured the growth rate by using commercial aluminium wires in each of which a crystal of a definite direction had been grown by an appropriate length. The rate was measured from the increase of length of the crystal during the successive annealing. Thus the rate was measured as a function of direction of the growing grain as well as of the temperature. They also measured G by means of travelling furnace method(4) in which the growth rate of the crystal was assumed to be equal with that of the

* The 729th report of the Research Institute for Iron, Steel and Other Metals.

(1) W. A. Anderson and R. F. Mehl, Trans. AIME, **161** (1945), 140.

(2) M. Kornfeld and W. Pawlow, Phys. Zeit, Sowjetunion, **12** (1937), 301.

(3) T. Fujiwara, *Butsurigaku Kôenshû*, **4** (Maruzen, Tokyo, 1944), 145 (in Japanese).

(4) T. Fujiwara, E. Tatsumoto and T. Hirokawa, J. Sci. Hiroshima Univ., **11** (1941), 311.

furnace. Interpolating the temperature at the head of the growing crystal, they obtained the rate of growth as a function of temperature and of crystal direction. The measurements were carried out on wires and on sheets[5]. Their results obtained on sheets differ markedly from those measurements mentioned above.

There are several investigations on the growth rate of recrystallized grain during primary recrystallization of other metals[6][7], alloys[8], and non-metallic materials[9], but these are not so thorough as those of aluminium.* It is desired at the present stage of the studies on recrystallization as well as on the nature of the grain boundary to perform more extensive measurements on the growth rate as a function of temperature, deformation, grain-size, impurity over a range of these variables as wider as possible. In this investigation, seven kinds of aluminium were used, those are pure aluminium and six kinds of Al-Cu alloys containing 0.027, 0.15, 0.56, 1.03, 1.61 and 2.00 wt.% copper, respectively, and the rates of growth were measured over the temperature range 296~622°C, for the deformation 1.2~15% in elongation on the specimens of grain-size 0.05~0.6 mm in the average diameter.

For simplicity the investigations by Anderson and Mehl, or Kornfeld and Pawlow will be henceforth referred to as A-M or K-P.

II. Experimental procedure

1. Preparation of specimens

The pure aluminium (99.995%)** supplied in the as-cast condition, was directly forged without remelting and then cold-rolled more than 95% to the thickness of about 0.4 mm. Other materials containing copper were produced by melting the pure aluminium in an alumina crucible adding a required amount of mother alloy, which had been prepared from the pure aluminium and from electrolytic copper (99.9%). These materials were casted in a shape of 4 cm × 4 cm in the cross section and of about 20 cm long. Those ingots were forged and then cold-rolled to about 0.4 mm thick. The materials are found by chemical analysis to contain 0.027, 0.15, 0.56, 1.03, 1.61, 2.00 wt.% copper, respectively.

Strips 2 cm wide and about 20 cm long were cut from those rolled materials with the long dimension parallel to the direction of rolling. After marking two parallel scratches parpendicular to the long dimension for the purpose to measure the amount of elongation given afterwards, these strips were annealed in order to provide uniform and equiaxed grain sizes. Table 1 denotes these annealing

(5) T. Fujiwara and T. Yamashita, J. Sci. Hiroshima Univ., **12** (1942), 163 (in Japanese).
(6) E. Tatsumoto, J. Sci. Hiroshima Univ., **12** (1942), 173 (in Japanese).
(7) R. Karnop and G. Sachs, Z. Phys., **60** (1930), 464.
(8) J. K. Stanley, Trans. AIME, **162** (1945), 116.
(9) H. G. Mülar, Z. Phys., **96** (1935), 279.

* An excellent review on this subject has recently been presented by Burke and Turnbull, *Progress in Metal Physics*, **III** (London, 1952), 220.

** By a preliminary spectroscopic analysis no traces except Mg and Cu were detected, Mg is perhaps of the order of 10^{-3}% and Cu is less than 10^{-3}%.

treatments. The materials were annealed at various temperatures from 300 to 600°C during 2 or 3 hours according to the desired grain size. The average value of grain diameters of an annealed material, D, is also given in the table. D is defined as the mean value of the lengths of line elements which are the parts of any straight line being cut by each grain boundary.

Table 1. Treatments of samples before cold-work.

Lot	Copper content in wt.%	Temperature of annealing in °C	Annealing time in hour	Average of grain diameter in mm
A I	0	300	2	0.057
A II	0	350	3	0.083
A III	0	350	2	0.11
A IV	0	400	2	0.15
A V	0	400	3	0.17
A VI	0	400	2	0.20
A VII	0	400	2	0.23
A VIII	0	500	2	0.27
A IX	0	600	2	0.34
A X	0	500	3	0.36
A XI	0	600	3	0.40
B I	0.027	400	2	0.16
B II	0.027	600	2	0.34
C I	0.15	400	2	0.16
C II	0.15	600	2	0.35
D I	0.56	400	2	0.14
D II	0.56	600	2	0.31
E I	1.03	350	2	0.093
E II	1.03	450	2	0.25
E III	1.03	550	2	0.63
F I	1.61	350	2	0.059
F II	1.61	450	2	0.26
G I	2.00	350	2	0.055
G II	2.00	450	2	0.26
G III	2.00	550	2	0.60

It was frequently observed in pure aluminium that there are a few grains with long dimensions as large as a few cm along the rolling direction but with very thin thickness being formed along the free surface of the specimen even during such a short time annealing as at 200°C for 30 minutes. These anomalous grains do not appear when the materials have been chemically etched to remove the surface layers about 0.05 mm thick before the annealing. It is striking that the anomalous growth was not observed in materials containing copper, even if its amount is so small as 0.027 wt.%.

It must be noted that D of Lot A IV, A VI and A VII, for example, does not agree with each other, in all of those cases the materials were annealed at the same temperature during the same time period. This fact seems to be caused from the dependence of D on the rate of heating as well as on the two variables above mentioned.

2. Treatments

The annealed strips were elongated in a simple tensile machine, which was specially designed to avoid any disturbance in attaching the material to the machine. As already emphasized by Anderson and Mehl[1], the elongation introduces very simple stress pattern in the sample, namely, no macroscopic stress originate, but only a uniformly distributed pattern of microscopic stress. The degree of elongation was measured by means of two gauge marks scratched before the annealing. Each strip provided five samples, the degree of elongation in the samples varying by not more than ± 0.1 per cent elongation.

The elongated samples were heated for recrystallization in a fused salt bath. Temperature variation within the bath amounted to ± 2°C.

It has been well established that the severer the deformation, the greater is the rate of growth, G, which is independent of annealing time periods. And it is also well known that the incubation time, in which a nuclei is formed in the sample, strongly depends on the degree of deformation, evidently, the shorter the period, the larger the amounts of deformation, as shown in Fig. 1.

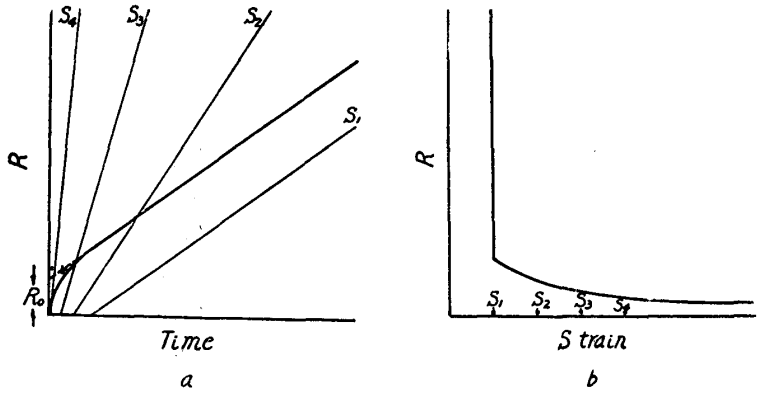

Fig. 1. (a) The thick curve is a schematic representation of the radius of a grain started from a needle indentation plotted against the time period of annealing. The thin lines are the plotts of radius of largest grain *vs.* time for the strains S_1, S_2, S_3 and S_4, respectively.
(b) Schematic representation of the stain distribution round the needle indentation.

Two or five points at the surface of a sample were pricked by a sharp needle, before the recrystallization, in order to accerelate the nucleation of the growing grains. When the sample is heated to recrystallize, a nucleus is readily formed at the needle indentation and is followed by rapid growth in the surrounding narrow region deformed severly by the prickling, then it grows in the region strained homogeneously by the previous elongation.

Then the observed values of the radii of growing grains will increase with the progression of annealing in accordance with the relation denoted by the thick curve in Fig. 1a. Namely, it may be supposed that a growing grain, started from the needle indentation, had a definite radius prior to the annealing and, as annealed, grows readily from this dimension at a constant rate corresponding to the strain previously supplied by the elongation. The rate of growth G, therefore, can be determined by plotting the observed radii of growing grains, started from the needle indentations, against the annealing time periods.

III. Time dependence of G

Fig. 2 shows an example of the measurement which was carried out at 506°C by using the samples of Lot AV elongated by 6.05 per cent. Here the vertical axis denotes the maximum radius of a grain started from the needle indentation in each of the samples, and the absissa is the annealing time. Those observed values seem to satisfy the relation denoted in Fig. 1a.

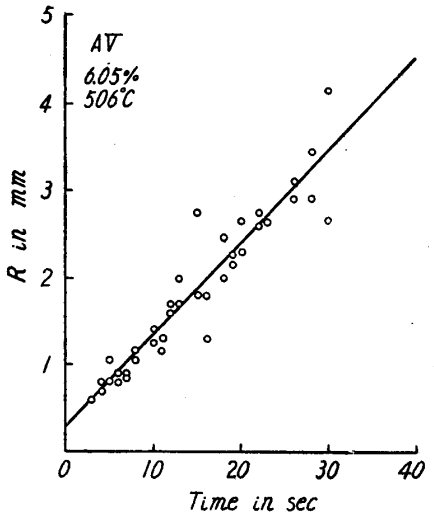

Fig. 2. Radius of a grain started from the needle indentation *vs.* time of annealing at 506°C, 6.05% elongation Lot AV aluminium.

However, it was sometimes found that the observed values fairly deviate from the relation of Fig. 1a. In the cases of low degree of deformation, the tangent of the curve denoting the radius against time decreases with increasing time. The radius of the growing grain seems to approach to a definite value, instead of to increase steadily, after a long time period as shown in Fig. 3a. This result is inconsistent with A-M's data. Hence, there arises the question as to whether the decrease of tangent denotes the true decrease of growth rate with increasing time, or merely an apparent change caused from the inhomogeneous strain distribution by the prickling. In these samples, however, a nucleus can also start from another point remote from the needle indentation and grows at a rate of the same order of magnitude with the grain started from the indentation. It is, therefore, concluded that the rate of growth G decreases with the increase of time period of annealing, even before the impingement of the growing grain with another takes place.

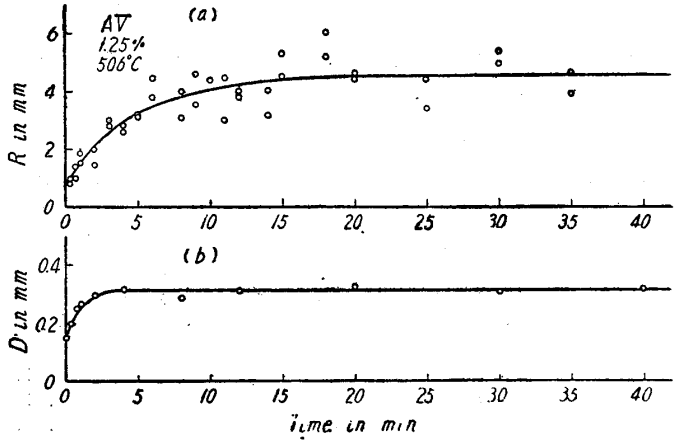

Fig. 3. (a) Radius of grain started from the needle indentation *vs.* time of annealing at 506°C, 1.25% elongation Lot AV aluminium.
(b) Average diameter *vs.* time of annealing without any cold deformation at 506°C Lot AV aluminium.

It is always observed in those cases of low degrees of deformation that uniform grain growth takes place over the strained matrix, concurrently with the nucleation and growth of new grains. Fig. 3a denotes the measurement carried out at 506°C on samples of Lot AV, which had previously been annealed at 400°C for 2 hours, extended by 1.25 per cent. It is easily supposed that those samples of Lot AV, but being not subjected to any cold deformation, would also reveal grain growth

by the final annealing at 506°C, and Fig. 3b represents the observed grain growth. It is, therefore, reasonable to consider that the growth rate decreases on account of the uniform grain growth taking place over the whole unconsumed region, which would diminish the driving energy, viz. the strain energy and the grain boundary energy per unit volume.

The time dependence of G is very unfavourable for our purpose to measure the rate of growth of recrystallized grain under various circumstances. In this investigation, therefore, the values of G were always extrapolated to the instant of the begining of the annealing, because the extrapolated value will correspond to the driving force which has not been subjected to any relaxation. It may be natural to assume that the radius of the growing grain R increases obeying the following relation with increasing time t,

$$R = R_0 + G_0\tau_0\{1 - \exp(-t/\tau_0)\}\ , \qquad (1)$$

where R_0 is the effective radius of the region severly deformed by the prickling, G_0 the extrapolated value of G at $t = 0$, τ_0 the relaxation time of the decrease of the growth rate. The curve of Fig. 3a denotes the calculated values of R according to (1), substituting $R_0 = 7\times10^{-2}$ cm, $G_0 = 1.2\times10^{-3}$ cm sec^{-1} and $\tau_0 = 288$ sec. The value of G_0 can, therefore, be determined with sufficient accuracy by means of the whole data over long time interval.

The increase of D is also denoted by an analogous relation with (1) as

$$D = D_0 + 2v_0\tau_0'\{1-\exp(-t/\tau_0')\}\ , \qquad (2)$$

where D_0 is the average of grain diameter before the annealing, $2v_0$ the rate of increase of D at $t = 0$, τ_0' the relaxation time of the decrease of rate of grain growth. It is a matter of course that G_0 is far larger than v_0, but it is unfavourable that τ_0 is five times as large as τ_0', because we must then suppose that the driving force does decrease after the completion of the grain growth in unconsumed region. The strange feature concerning τ_0/τ_0', however, seems to be removed at sufficiently high temperatures, because the value τ_0/τ_0' decreases with the increase of temperature as denoted in the Table 2.

Table 2. Comparison of G_0 with v_0 and of τ_0 with τ_0' in Lot AV.

Temperature in °C	G_0 in cm sec^{-1}	τ_0 in sec	v_0 in cm sec^{-1}	τ_0' in sec	τ_0/τ_0'
506	1.2×10^{-3}	288	1.6×10^{-4}	52	5.5
574	8.1×10^{-3}	85	5.5×10^{-4}	20	4.3
622	2.4×10^{-2}	15	2.1×10^{-3}	7.4	2.0

The large values of τ_0/τ_0' at rather low temperatures will presumally be caused from the coasening of polygonization structures as suggested in another paper[10]. Any further confirmations on this prediction were, however, not given by means of X-ray or microscopic examination. Whether the suggestion may be true or not, the discordancy between A-M's results and our own will be accounted on the

(10) H. Suzuki, To be published in this journal.

basis of the large values of τ_0/τ_0' at low temperatures. Sine τ_0' seems to be proportional to the time periods required to the measurements, τ_0 would be far larger than the time periods of A-M's measurements at such low temperatures as they investigated.

IV. Results of measurements

The extrapolated value of rate of growth at $t = 0$ can now be computed from the plot R *vs.* t observed under various conditions. The results thus obtained are represented in Table 3, where D is the average of grain diameter before elongation, l the degree of elongation and G_0 the extrapolated rate of growth of recrystallized grain at $t = 0$.

Table 3. Results of measurements.

Temperature in °C	D in mm	l in %	G_0 in cm sec^{-1}	Temperature in °C	D in mm	l in %	G_0 in cm sec^{-1}
Pure Al							
622	0.17	1.25	2.4×10^{-2}	526	0.34	3.17	1.0×10^{-3}
	0.23	2.84	4.3×10^{-2}			4.15	2.8 "
		3.52	5.5 "			6.15	6.0 "
		4.11	6.6 "			8.16	1.0×10^{-2}
	0.34	2.50	1.7×10^{-2}		0.36	4.00	2.1×10^{-3}
		3.48	2.6 "			6.05	5.5 "
		4.55	5.2 "			8.80	7.4 "
		5.50	6.5 "		0.46	4.00	1.2×10^{-3}
574	0.17	1.25	8.1×10^{-3}			6.05	2.7 "
						8.80	4.2 "
	0.23	1.95	5.8×10^{-3}	506	0.17	1.25	1.2×10^{-3}
		2.49	1.1×10^{-2}			2.60	3.6 "
		3.33	1.8 "			4.05	6.0 "
		4.78	3.5 "			6.05	1.1×10^{-2}
		5.30	3.8 "			8.80	1.4 "
	0.34	3.14	1.0×10^{-2}	478	0.23	3.53	8.3×10^{-4}
		3.98	1.6 "			4.77	1.5×10^{-3}
		5.15	2.1 "			6.81	3.2 "
		5.93	3.5 "				
526	0.083	1.25	7.6×10^{-3}	428	0.057	2.15	4.4×10^{-4}
		2.50	1.6×10^{-2}			3.06	8.5 "
		4.00	1.9 "			4.20	1.6×10^{-3}
		6.05	2.0 "			5.20	2.4 "
						6.34	2.4 "
	0.11	2.16	1.2×10^{-2}		0.11	2.21	1.3×10^{-4}
		3.04	1.3 "			3.77	2.7 "
		4.22	1.7 "			5.08	6.8 "
		5.13	2.1 "			6.03	1.1×10^{-3}
						7.57	1.3 "
	0.17	1.25	1.8×10^{-3}		0.23	3.92	5.3×10^{-5}
		2.50	5.0 "			4.52	9.8 "
		4.00	9.0 "			5.40	1.2×10^{-4}
		6.05	1.2×10^{-2}			7.42	4.3 "
	0.23	3.18	3.8×10^{-3}		0.34	4.32	1.2×10^{-5}
		4.23	7.0 "			5.69	3.3 "
		5.02	8.5 "			8.28	6.0 "
		6.49	1.1×10^{-2}				

Table 3. Continue

Temperature in °C	D in mm	l in %	G_0 in cm sec^{-1}
Pure Al (cont.)			
378	0.23	5.24	2.6×10^{-5}
		6.40	2.7 "
		7.17	3.8 "
		8.33	4.8 "
350	0.10	3.95	4.2×10^{-6}
		6.05	1.3×10^{-5}
		8.63	1.9 "
	0.17	6.12	3.4×10^{-6}
0.027% Cu			
622	0.16	1.42	2.5×10^{-2}
		1.96	4.4 "
		2.55	7.2 "
		3.89	9.8 "
	0.34	3.32	4.0×10^{-2}
		4.04	4.8 "
574	0.16	1.57	9.6×10^{-3}
		2.58	2.4×10^{-2}
		4.14	5.4 "
526	0.16	1.63	1.0×10^{-3}
		3.02	7.3 "
		4.79	1.6×10^{-2}
		6.65	3.0 "
	0.34	3.14	4.8×10^{-4}
		5.08	3.0×10^{-3}
		6.87	6.2 "
477	0.16	1.72	1.9×10^{-4}
		2.55	1.0×10^{-3}
		4.05	1.9 "
		6.75	4.9 "
428	0.16	1.70	2.7×10^{-5}
		2.93	6.2 "
		4.07	2.0×10^{-4}
		6.27	4.0 "
		8.07	7.0 "
378	0.16	2.47	8.3×10^{-7}
		3.37	1.6×10^{-6}
		4.15	7.1 "
		6.14	1.7×10^{-5}
0.15% Cu			
622	0.16	1.35	2.2×10^{-2}
		1.99	3.2 "
		2.75	5.5 "
		3.95	8.4 "
	0.35	2.12	1.0×10^{-2}
		3.18	2.5 "
		4.84	5.5 "
574	0.16	1.59	9.6×10^{-3}
		2.72	2.6×10^{-2}
		4.12	4.5 "
526	0.16	1.63	2.8×10^{-3}
		2.68	6.8 "
		4.70	1.4×10^{-2}
		6.71	1.8 "
		9.8	2.5 "
	0.35	3.33	1.1×10^{-3}
		4.52	2.7 "
		6.58	4.5 "
477	0.16	1.75	3.1×10^{-4}
		2.73	1.2×10^{-3}
		4.55	3.0 "
		6.31	4.5 "
		8.16	6.0 "
428	0.16	1.53	9.2×10^{-6}
		2.66	1.3×10^{-4}
		4.01	2.8 "
		6.41	6.3 "
		8.9	1.1×10^{-3}
	0.35	4.63	8.3×10^{-6}
		5.32	2.3×10^{-5}
		6.56	7.2 "
		8.43	1.5×10^{-4}
378	0.16	2.82	1.6×10^{-6}
		4.12	1.3×10^{-5}
		6.33	3.4 "
		8.40	9.3 "
0.56% Cu			
622	0.14	1.35	1.3×10^{-2}
		2.58	2.7 "
		3.09	2.8 "
		3.99	4.1 "
	0.31	1.63	5×10^{-3}
		2.74	1.4×10^{-2}
		3.82	2.2 "
574	0.14	1.47	4.0×10^{-3}
		2.60	9.5 "
		3.50	1.8×10^{-2}
		5.31	2.9 "
526	0.14	1.43	5.7×10^{-4}
		2.61	3.1×10^{-3}
		3.57	5.3 "
		5.08	8.0 "
		7.22	1.1×10^{-2}
	0.31	2.21	1.8×10^{-4}
		3.16	5.7 "
		4.83	2.3×10^{-3}
		6.95	4.4 "

Table 3. Continue

Temperature in °C	D in mm	l in %	G_0 in cm sec^{-1}
477	0.14	1.42	$1.1\times10^{-}$
		2.29	5.3 "
		3.59	1.1×10^{-3}
		5.62	2.7 "
		8.39	3.7 "
428	0.14	1.48	1.4×10^{-5}
		2.89	1.0×10^{-4}
		3.69	1.5 "
		5.58	5.0 "
		7.50	6.7 "
	0.31	4.09	1.5×10^{-5}
		5.64	5.8 "
		6.13	8.5 "
		7.74	1.3×10^{-4}
378	0.14	3.64	1.7×10^{-5}
		5.59	2.6 "
		6.79	4.8 "
		8.87	7.5 "
327	0.14	3.51	4.4×10^{-7}
		4.84	1.4×10^{-6}
		6.56	2.5 "
		8.05	3.9 "
1.03% Cu			
596	0.25	1.79	3.4×10^{-3}
		3.06	8.5 "
		4.63	1.3×10^{-2}
	0.63	3.07	3.1×10^{-3}
		4.08	4.2 "
		5.59	5.6 "
547	0.25	1.76	1.6×10^{-3}
		3.05	3.3 "
		4.58	6.2 "
		6.04	8.7 "
	0.63	3.34	8.8×10^{-4}
		4.21	1.5×10^{-3}
		6.30	2.5 "
498	0.25	1.85	3.8×10^{-4}
		3.04	1.3×10^{-3}
		4.59	2.1 "
		6.03	2.3 "
449	0.093	0.74	2.5×10^{-4}
		1.17	5.4 "
		1.91	1.0×10^{-3}
		3.13	1.6 "
	0.25	1.80	3.7×10^{-5}
		3.03	$2.1\sim3.0\times10^{-4}$
		4.50	$4.6\sim5.8$ "
		5.98	9.0 "
		7.69	1.2×10^{-3}
400	0.093	0.71	3.2×10^{-5}
		1.16	6.5 "
		1.84	1.5×10^{-4}
		3.00	2.1 "
	0.25	3.12	2.6×10^{-5}
		4.55	7.0 "

Temperature in °C	D in mm	l in %	G_0 in cm sec^{-1}
1.61% Cu			
596	0.26	1.82	2.2×10^{-3}
		3.03	4.4 "
		4.47	8 "
547	0.26	1.86	1.0×10^{-3}
		3.01	2.7 "
		4.55	4.8 "
		5.96	6 "
498	0.26	1.8	4.6×10^{-4}
		3.05	1.3×10^{-3}
		4.47	2 "
		6.05	2.6 "
449	0.059	1.00	1.2×10^{-4}
		1.86	2.5 "
		3.06	7.0 "
	0.26	1.76	7.6×10^{-5}
		2.95	2.9×10^{-4}
		4.46	5.5 "
		6.03	7 "
		8.02	8 "
2.00% Cu			
596	0.26	2.97	4×10^{-3}
		4.45	7.5 "
	0.60	2.80	1.2×10^{-3}
		3.98	1.9 "
		5.76	3.0 "
547	0.26	1.68	6.8×10^{-4}
		2.94	2.2×10^{-3}
		4.42	4 "
		6.00	7 "
	0.60	3.14	6×10^{-4}
		4.50	1.4×10^{-3}
		6.06	2 "
498	0.26	1.66	3.5×10^{-4}
		3.12	1.3×10^{-3}
		4.55	2.1 "
		5.95	2.7 "
449	0.055	1.05	1.3×10^{-4}
		1.77	2 "
		3.21	3.1 "
		4.62	7.5 "
	0.26	2.92	2.7×10^{-4}
		4.46	5.6 "
		5.96	8.8 "
		8.12	1.3×10^{-3}
399	0.055	4.50	1.2×10^{-4}

V. Effects of initial grain size and degree of deformation

1. Pure aluminium

It has been known since Anderson and Mehl's measurement that the larger the initial grain size, the smaller is the rate of growth G. However there has not been established any quantitative relation between G and D as yet.

Fig. 4a represents an example of measurement of G_0 carried out by using pure aluminium with various grain sizes at 526°C, where $\log G_0$ is plotted against $\log l$.

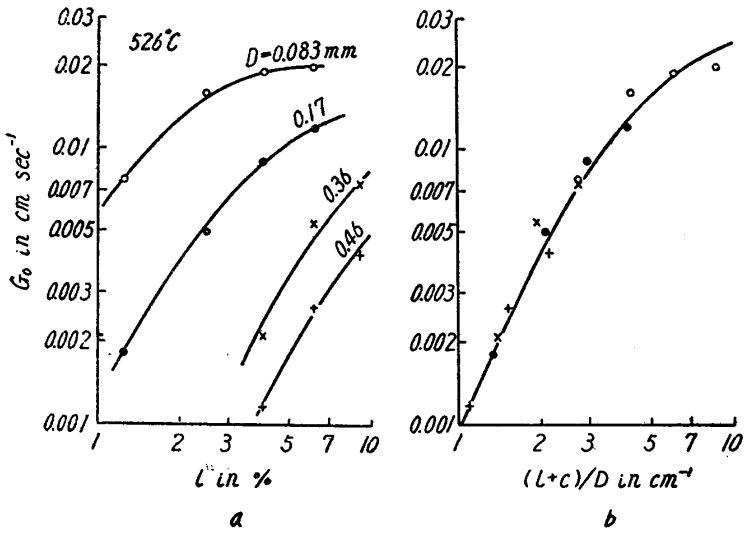

Fig. 4. (a) Log G_0 *vs.* log l at 526°C, Lot A.
(b) The same quantities with (a) plotted against $\log\{(l+c)/D\}$.

As already known, the rate of growth decreases with increasing grain size, when the degree of deformation is fixed at a constant value. It is striking that the values of $\log G_0$, represented in Fig. 4a can be superposed on only one curve by plotting against $\log\{(l+c)/D\}$ instead of $\log l$ as shown in Fig. 4b. Here c is a constant with the magnitude of 1.02 per cent. This relation remains true in the measurements carried out at the temperatures below 540°C on pure aluminium. Fig. 5 denotes the values of $\log G_0$, measured at various temperature, plotted against $\log\{(l+c)/D\}$. As easily seen from this figure the relation mentioned above fails at temperatures higher than 570°C, namely, the observed values of $\log G_0$ at the same temperature do not form only one curve, when these are plotted against $\log\{(l+c)/D\}$. In the high temperature region G_0 is rather a function of l/D as seen from Fig. 6.

It is interest to note that the already published data measured by using samples with various grain sizes also obey the above mentioned rule.

Of course, the abscissa of Fig. 5 or 6 denotes a quantity corresponding to the driving force of the growth of recrystallized grain during the primary recrystallization. The driving force is therefore a function of $\log\{(l+c)/D\}$ at temperatures below 540°C. It is easily shown that the released grain boundary energy during the growth of a grain consuming the matrix grains is given by BE/D[10], where B is a constant with the magnitude of 1.6~1.9, and E the average of grain boundary energies per unit area. Consequently the driving energy U is given by the expression

$$U = BE(l + c)/Dc \ , \qquad (3)$$

or

$$F = B\sigma(l + c)/Dc \ , \qquad (4)$$

where F is the driving force and σ the average of grain boundary tensions. It seems, therefore, to be strange that the driving force is expressed only in terms of l/D at the higher than 570°C. Discussions concerning this matter will appear in another paper[10] concurrently with a quantitative treatment of the driving force.

According to the above arguments, it is now able to estimate the absolute magnitude of the driving energy for the growth of recrystallized grain. Assuming $E = 400\,\mathrm{erg\,cm^{-2}}$, we have $BE/C \simeq 7\times10^{4}\,\mathrm{erg\,cm^{-2}}$, which corresponds to the driving energy $7\times10^{4}\,\mathrm{erg\,cm^{-3}}$ or $6\times10^{-4}\,\mathrm{cal\,g^{-1}}$ in the case of $(l+c)/D = 1\,\mathrm{cm^{-1}}$, and to 6×10^{-3} cal $\mathrm{g^{-1}}$ in the case of $(l+c)/D = 10\,\mathrm{cm^{-1}}$. These values of energy are far smaller than that expected from the stored energy during cold deformation. The circumstances are compatible with the measurement by Taira Suzuki[11] on release of stored energy during cold work, in which he found by measuring the specific heat of cold worked copper that the most part of the stored energy had been released before the recrystallization being observed.

It is seen from Figs. 5 and 6 that G_0 is a complicated function of F. If F is small, G_0 is proportional to F^n, where n takes a value varying from 2 to 4 at temperatures lower than 540°C. n decreases with increasing F. At temperatures higher than 570°C, G_0 increases at the rate of 1~1.5 th power of l/D.

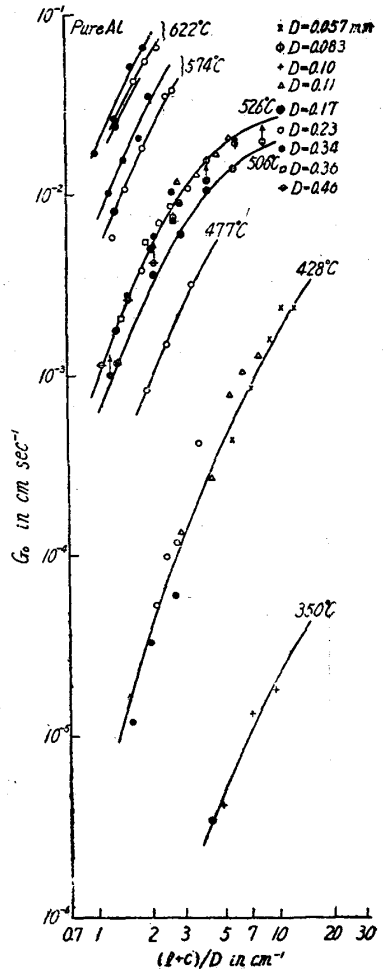

Fig. 5. Log G_0 *vs.* log$\{(l+c)/D\}$, Lot A.

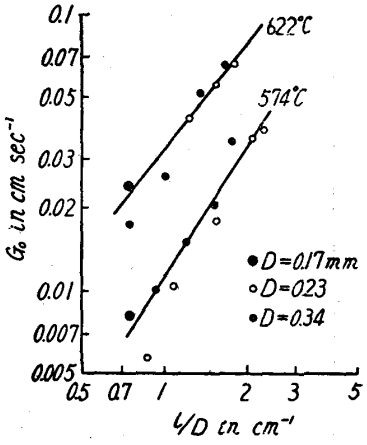

Fig. 6. Log G_0 *vs.* log (l/D), Lot A.

Some measurements were also carried out at rather low temperatures by an abbreviate method using Lot A IV. Samples with 2 cm wide and about 7 cm long were pricked by a needle at five points in each of them after supplying required amounts of elongation by the tensile machine, then were recrystallized at the temperature of measurement. G_0 is determined by each of samples, under the assumption that $R_0 = 0.3$ mm, and $\tau_0 \gg t$ in the relation (1). Fig. 7 represents log G_0 plotted against log l obtained by this abbreviate method.

2. Al-Cu alloys

Figs. 8, 9, 10 and 11 denote log G_0 plotted against log $\{(l+c)/D\}$ in the cases

(11) T. Suzuki, Sci. Rep. RITU, **A1** (1949), 55.

of Al-Cu alloys which involve 0.027, 0.15, 0.56, and 1.03 per cent copper, respectively. Figs. 12 and 13 denote $\log G_0$ plotted against $\log (l/D)$ of alloys of 1.61 and 2.00 per cent copper, respectively. Though the measurements were not so satisfactorily performed over various grain sizes as in the case of pure aluminium, it is seen from these figures that G_0 can not be represented as an unique function of $(l+c)/D$ at the higher than a temperature which is characteristic to the copper content. At higher temperatures than those, the observed values of G_0 form a curve irrespective to the grain size, if these are plotted against l/D, except the cases of small values of l.

Curves in Figs. 8~13 resemble to those in Fig. 6 or 7, which represents the observation on pure aluminium, but a significant difference is observed between the measurements carried out by using Lot F I and F II or G I and G II. G_0

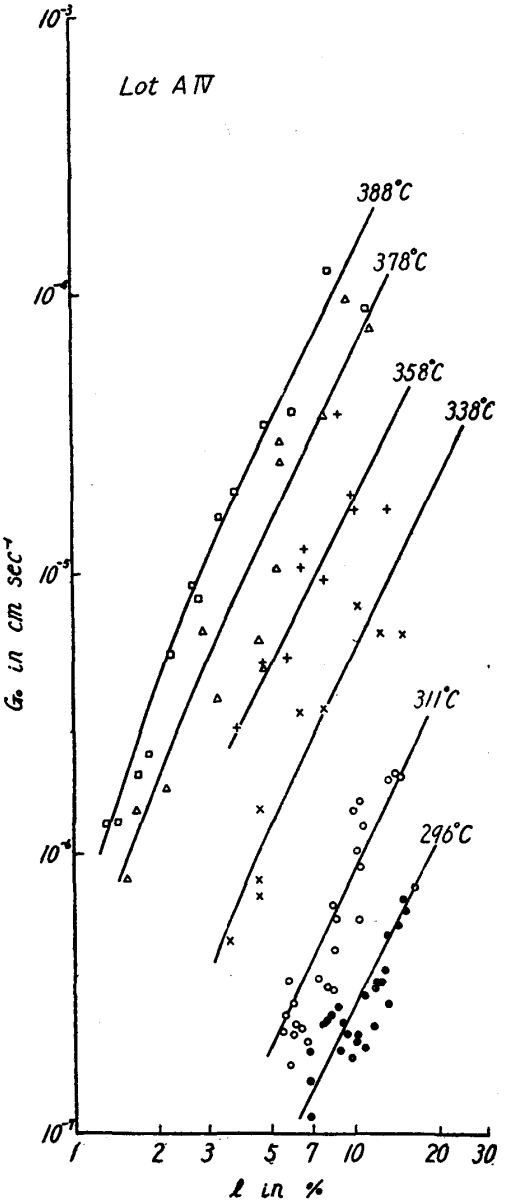

Fig. 7. Log G_0 *vs.* log l, Lot A IV.

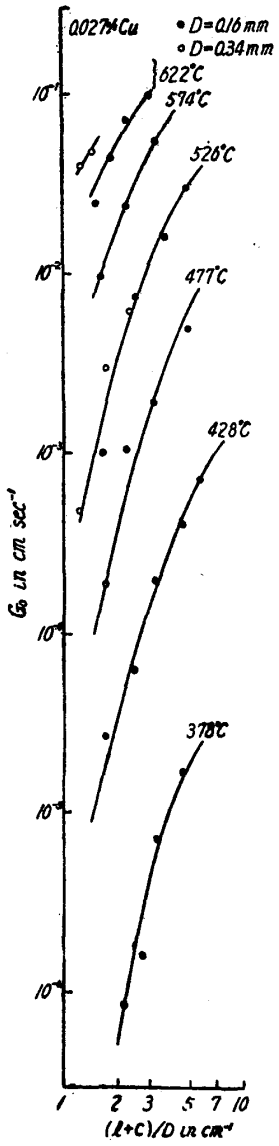

Fig. 8. Log G_0 *vs.* log$\{(l+c)/D\}$, Lot B.

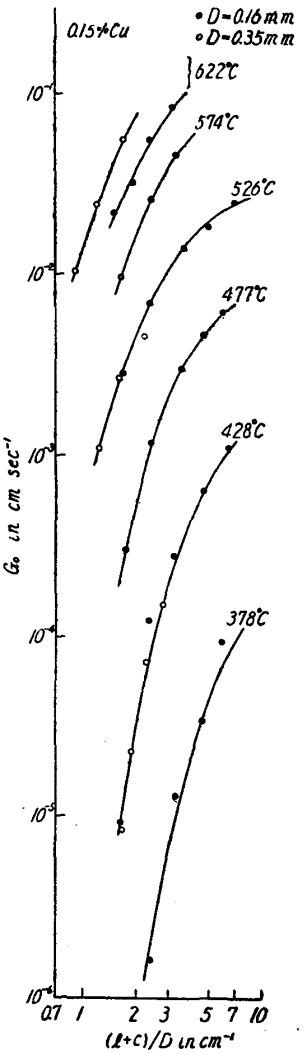

Fig. 9. Log G_0 *vs.* log$\{(l+c)/D\}$, Lot C.

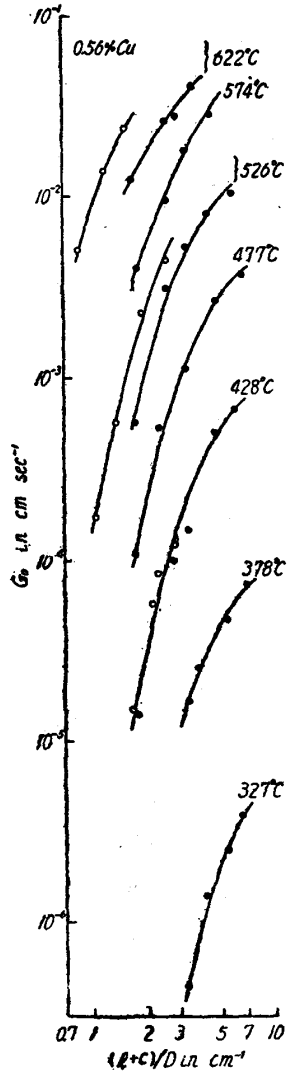

Fig. 10. Log G_0 *vs.* log $\{(l+c)/D\}$, Lot D.

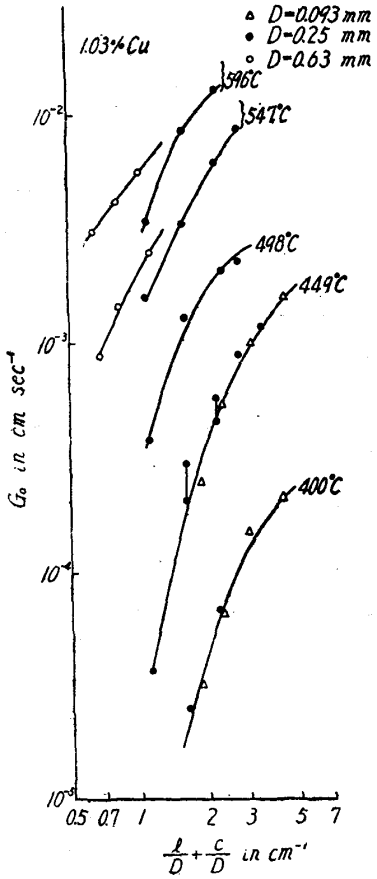

Fig. 11. Log G_0 *vs.* log $\{(l+c)/D\}$, Lot E.

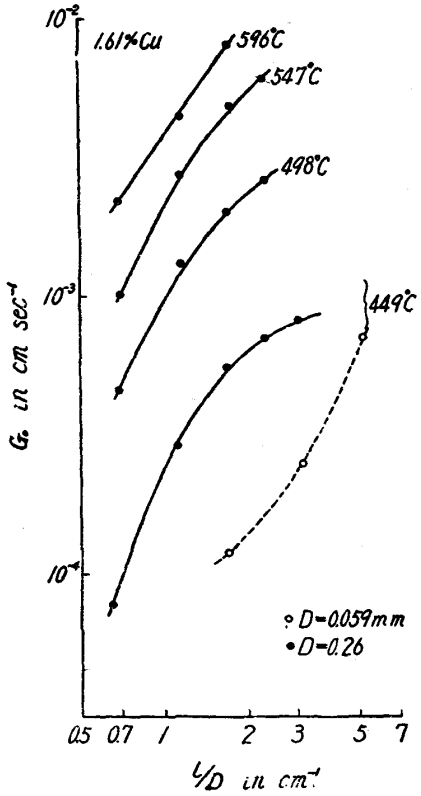

Fig. 12. Log G_0 *vs.* log (l/D), Lot F.

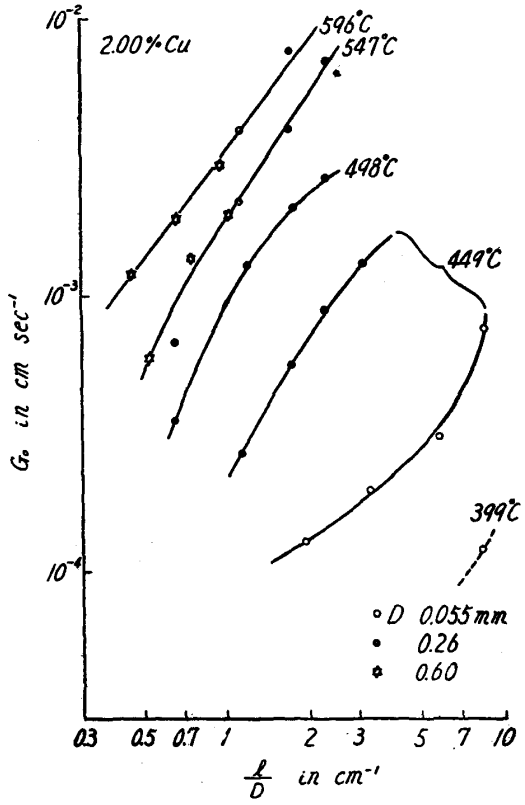

Fig. 13. Log G_0 *vs.* log (l/D) Lot G.

observed in the samples of Lot F I (or G I) are far smaller than those of Lot F II (or G II), even if the observed values of G_0 are plotted against $(l+c)/D$ as well as l/D. As already shown in Table 1 Lots F I, F II, G I and G II contain 1.61 or 2.00 per cent copper, and were annealed previously at 350 and 450°C, respectively. Therefore the precipitation of secondary phase should take place in samples of Lot F I or G I during the annealing previously subjected, while in Lot F II or G II the precipitation would scarcely occur. Consequently, it may be concluded that the precipitates make to retard considerably the growth of recrystallized grain on account of their distribution in matrix as well as of their dissolution.

VI. Effects of temperature on G_0

1. Pure aluminium

According to the considerations in the previous section the temperature dependence of G_0 is now able to be examined under the prescription of the same driving force. Fig. 14 denotes $\log G_0$ plotted against the reciprocal of the absolute temperature, taking the driving force as the perameter, namely, the curves are drawn for the fixed values of $(l+c)/D$ or l/D according to whether the temperature region is higher than 540°C or lower than this, respectively. In this figure the usual plots are also denoted by broken curves, which correspond to samples with $D = 0.20$ mm.

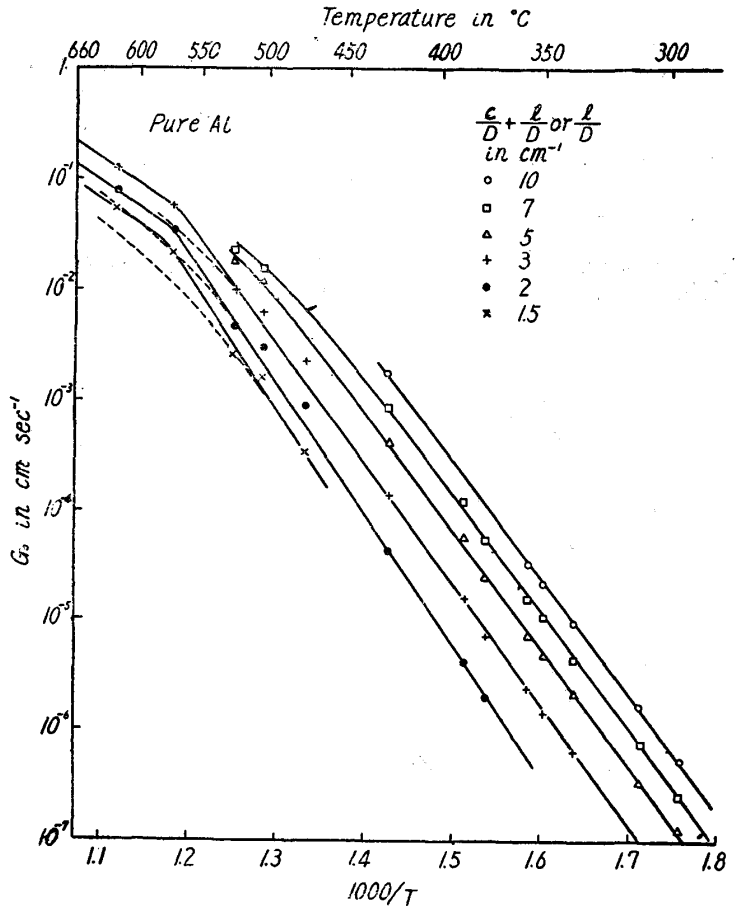

Fig. 14. Log G_0 *vs.* $1/T$ Lot A. The full curves are drawn for the fixed values of driving force, while the broken curves are the usual plot fixing the grain size and the degree of deformation.

It must be mentioned that both of the plots $\log G_0$ *vs.* $1/T$ in Fig. 14 do not reveal straight lines over the entire temperature range of the measurements but form concaved curves to the downwards, as Burke and Turnbull[12] have suggested by combining K-P's and A-M's data. The plots, however, form straight lines in the temperature range below 400°C, being compatible with A-M's data. But the slopes of the lines differ markedly from A-M's results.

(12) J.E. Burke and D. Turnbull, *Progress in Metal Physics*, **III** (London, 1952), 220.

2. Al-Cu alloys

The plot $\log G_0$ *vs.* $1/T$ for a fixed value of the driving force does not exhibit a straight line, but forms a curve concaved to downwards also in cases of Al-Cu alloys. Figs. 15~20 show the plots $\log G_0$ *vs.* $1/T$, taking the driving force as the parameter in the cases of alloys of 0.027, 0.15, 0.56, 1.03, 1.61 and 2.00 per cent Cu, respectively. As easily seen from these figures the slopes of the curves decrease with increasing copper content when the amounts of copper are more than 0.15 per cent. It is, however, found from Fig. 14, 15 and 16 that the slope decreases with further decrease of copper content less than 0.03 per cent.

3. Activation energies at low temperature range

It is obvious that we can not obtain the knowledge concerning the activation energy or entropy directly from the data without any theoretical considerations concerning the atomistic mechanism of the grain boundary migration, because the rate is not proportional to $\sinh(-F\lambda/kT)$ as well as the plot $\log G_0$ *vs.* $1/T$ does not form a straight line, where λ is the so-called reaction path. However, the activation energy computed by usual procedure may perhaps have a significance for rather low temperatures at which $\log G_0$ plotted against $1/T$ forms a straight line.

The activation energies thus computed are plotted against $(l+c)/D$ in Fig. 23, in which A-M's data are also drawn assuming $D = 0.056$ mm. The activation energy for the smallest driving force is considerably greater than others in almost cases, but our

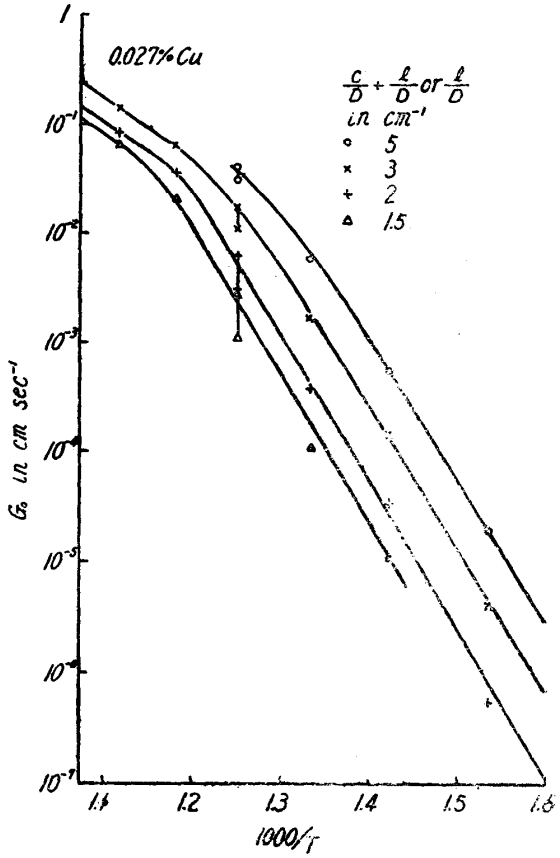

Fig. 15. $\log G_0$ *vs.* $1/T$ Lot B.

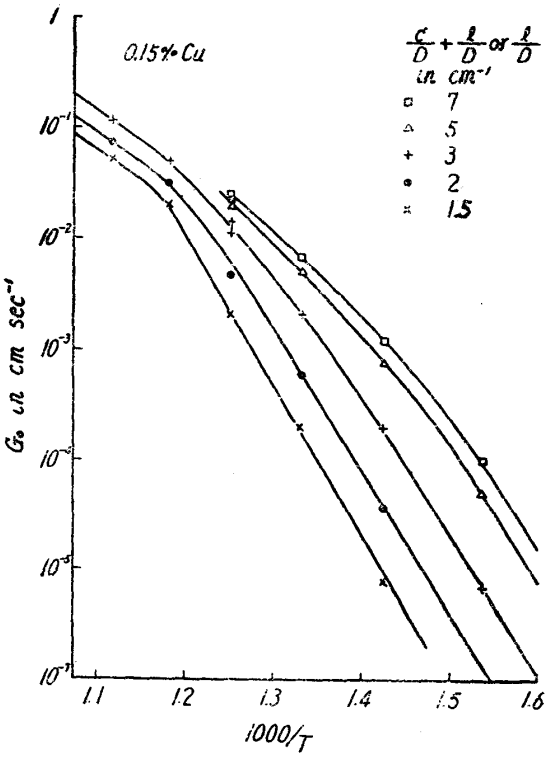

Fig. 16. $\log G_0$ *vs.* $1/T$ Lot C.

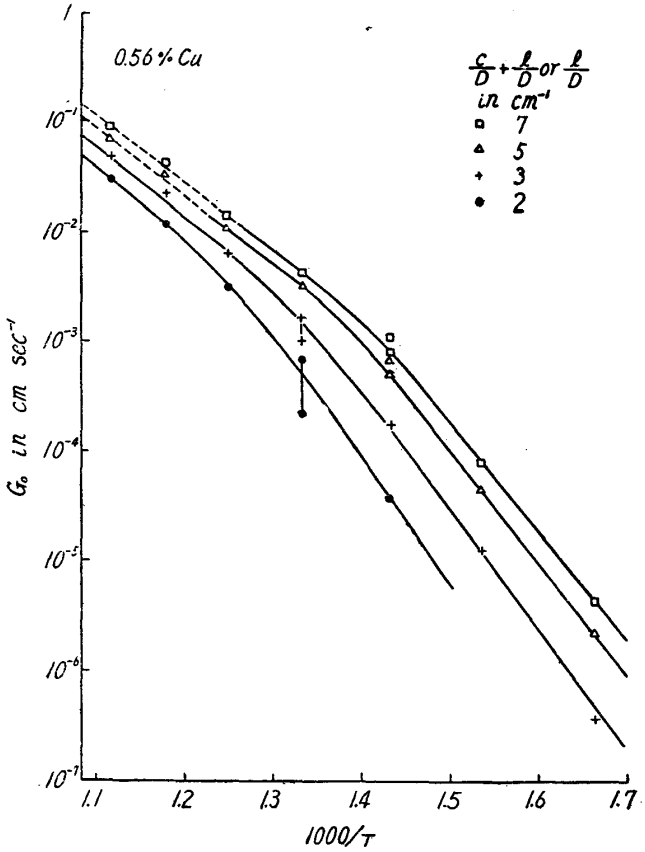

Fig. 17. Log G_0 *vs.* $1/T$ Lot D.

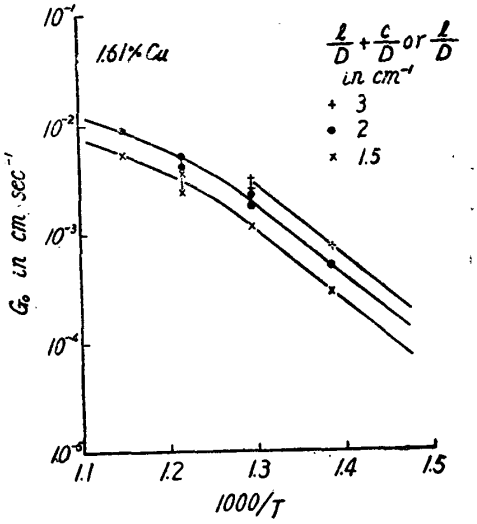

Fig. 19. Log G_0 *vs.* $1/T$ Lot F.

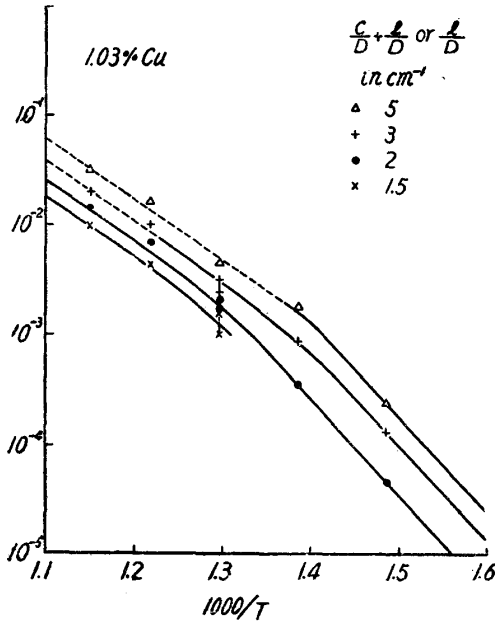

Fig. 18. Log G_0 *vs.* $1/T$ Lot E.

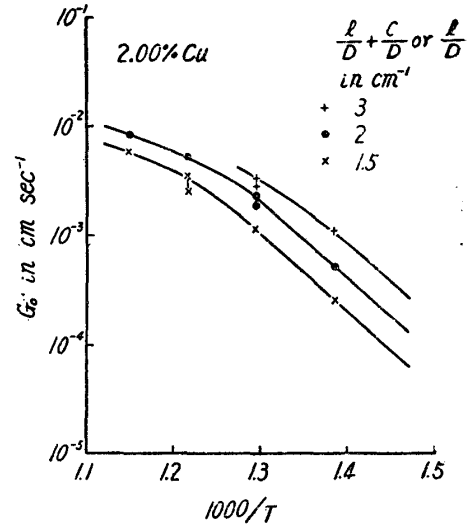

Fig. 20. Log G_0 *vs.* $1/T$ Lot G.

measurements were not so accurate as to draw any conclusions concerning the dependence of activation energy upon the driving force from Fig. 21. It is, however, striking that the activation energy reveals the maximum corresponding to the purity about 99.9 per cent as shown in Fig. 22, which shows the activation

energy plotted against the purity of samples. It has been usually supposed that the activation energy increases with increasing purity of specimens. But recently Chossat[13] have found that the recrystallization temperature of 99.998 per cent aluminium is remarkably lower than 99.99 per cent aluminium, and Kawashima and Nakamura[14] have found that the activation energy of the recrystallization sharply decreases when the purity of samples increases from 99.992 to 99.998 per cent. These results may reasonably be explained on the basis of our data.

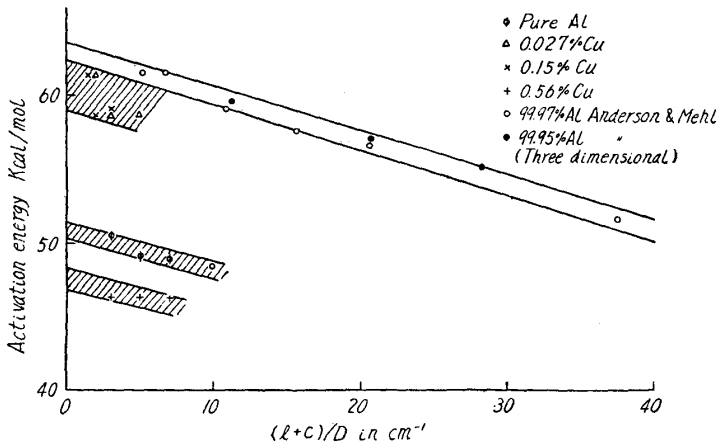

Fig. 21. Activation energy *vs.* $(l+c)/D$.

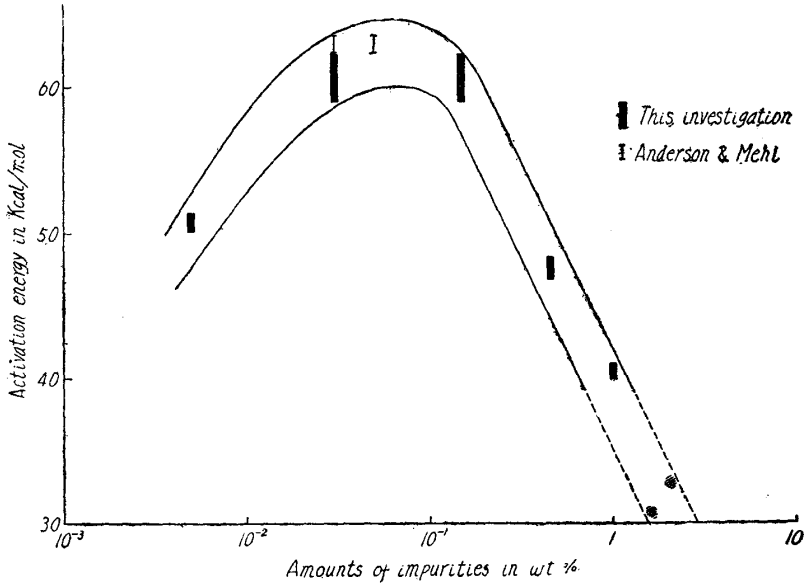

Fig. 22. Activation energy *vs.* amount of impurities.

Some theoretical difficulties concerning the extremely large activation energy in A-M's experiment may be removed by the concentration dependence of the activation energy mentioned above, because the energy of perfectly pure samples would be far smaller than those by them. Namely, it is suggested that a part of the exclusively large activation energy of A-M's measurement seems to be required for the associated motion of impurity atoms which form a kind of atomosphere round the grain boundary. The migration of the purely geometrical imperfections at the grain boundary requires probably a rather small activation energy comparable with the self-diffusion.

VII. Comparison with the other investigations

Fig. 23 denotes the available data, previously published, on the growth rate of recrystallized grain during the primary recrystallization of aluminium. A-M used

(13) H. Chossat, Rev. de Met., **47** (1950), 306.
(14) N. Kawashima and Y. Nakamura, J. Japan Inst. of Metals, **16** (1952), 183.

high purity aluminium (99.97 per cent) in their investigation. Therefore it is a matter of course that the activation energy in their measurement is almost equal to that of Lot B, which contains 0.027 per cent copper. The magnitudes of G_0 in their measurements also reveal good agreement with the results of Lot B, assuming D of A-M's samples to be 0.056 mm. Since the grain size of their samples was reported as 530 mm^{-2}, the assumed value of D seems to be reasonabe.

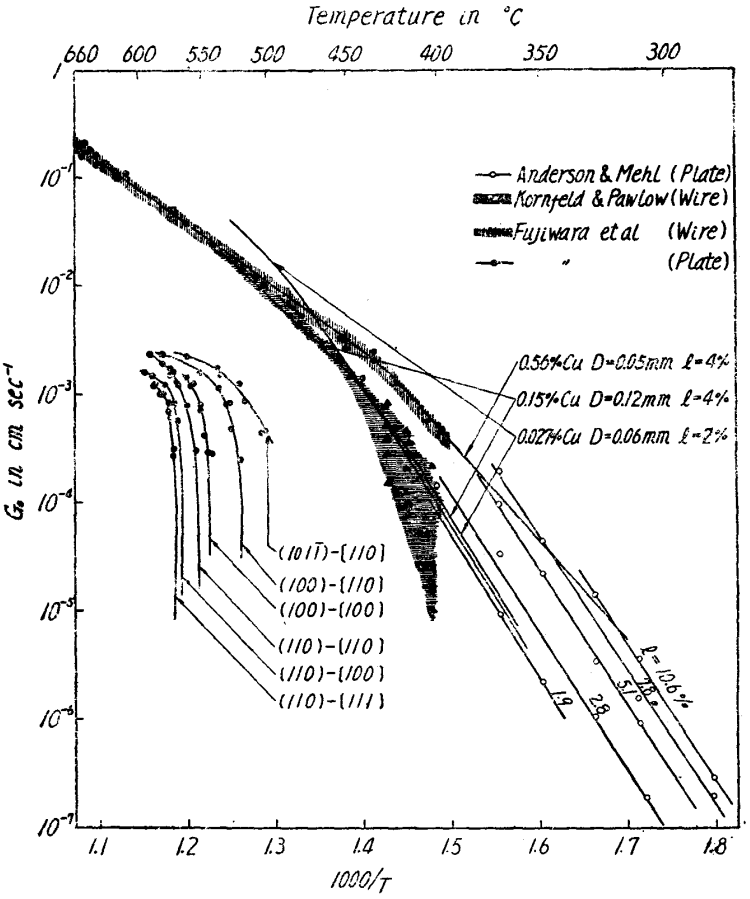

Fig. 23. Comparism with the available data of the growth rate in aluminium.

The activation energy evaluated from K-P's data is approximately 34 Kcal/mol, which differs markedly from A-M's value, i.e. 56~62 Kcal/mol. The reason of this divergence is now explained on the basis of this investigation. It is caused in a part from the greater amount of impurity in K-P's specimen than that in A-M's one, and in another part from the difference of temperature range over which the measurements were carried out. The failure of straight line relationship between log G_0 and $1/T$ is also found in K-P's data at temperatures below 450°C.

The results obtained by Fujiwara resemble qualitatively with those of K-P, as shown in Fg. 23. But the activation energy obtained at higher temperatures than 450°C is smaller than K-P's value determined over the same temperature range. The difference seems to be caused from the larger amount of impurity in samples used by Fujiwara than K-P's one.

Three curves, which fit with those results above mentioned, are extrapolated from the data in this investigation, and are drawn in the figure. These curves are drawn for samples of the same grain size stretched by the same amount, but not for the same driving force. Fujiwara's data fit the curve for the samples containing 0.56 per cent Cu with the grain size 0.05 mm in the average diameter and stretched by 4 per cent. The curve is extrapolated by means of Fig. 10 and of a similar figure which denotes log G_0 plotted against l/D. The measurements by K-P agree with the curve corresponding to the samples of 0.15 per cent Cu

with the grain size 0.12 mm in average diameter and elongated by 4 per cent. The results obtained by using the samples of 0.027 per cent Cu reveal sufficient agreement with A-M's data as already mentioned.

Fujiwara and Yamashita's data obtained by using aluminium sheets reveal very complicated dependence on temperature. Their results are shown in the left hand lower side of Fig. 23, each of the curves corresponds to a definite direction of growing grains. The failure of linear relationship between $\log G$ and $1/T$ is very pronounced in this case, namely, G falls sharply to zero as the temperature decreases to the characteristic one which depends on the direction of the growing grain. It must be remembered that in their investigation the crystal grows always consuming the matrix grains which have been heated very slowly to the growing temperature depending on the travelling rate and temperature distribution of the furnace. Then it is easily supposed that the uniform grain growth would occur over the strained matrix as mentioned in section III, because the samples had been stretched by 3.5 per cent after the pre-annealing at 500°C for 5 hours and then were heated to the temperatures above 500°C. The growth rates observed by Fujiwara and Yamashita should, therefore, be slower than the other investigations. Particularly, the growth rates of samples denoted in Fig. 3a would be zero, if it were measured by means of the travelling furnace method provided that the temperature gradient of the furnace is not extremely large. In accordance with this inference, the growth of new crystal in a wire also could entirely not be observed at temperatures below a critical one by means of the travelling furnace method. While the growth rates of the same samples measured by the isothermal method, in which the samples were rapidly heated to the annealing temperature by a salt bath, were comparable with those of the other investigations.

The apparent divergences between the exsisting investigations are, therefore, reasonablly interpreted on the basis of this investigation.

Summary

The growth rates of recrystallized grains in cold worked aluminium plates were measured by the method employed by Anderson and Mehl with slight modification. The measurements were carried out on seven kinds of aluminium under various circumstances, namely, on pure aluminium (99.995 per cent), Al-Cu alloys involving 0.027, 0.15, 0.56, 1.03, 1.61, 2.00 wt. per cent Cu, with the grain size 0.05~0.6 mm in average diameter, elongated by 1.2~15 per cent and at 296~622°C. The results are obtained as follows.

(1) The growth rate of a recrystallized grain decreases with increasing time period of annealing, even before the impingement of the growing grain with another takes place. This tendency is pronounced in samples of low degrees of deformation and at high temperatures.

(2) There are important relations between the rate of growth and the initial grain size, namely, the growth rate is a function of $(l+c)/D$ at a temperature below the characteristic one, which depends on the purity of the samples, and is

a function of l/D at the higher than this, approximately. Here l is the degree of elongation, c a constant with the magnitude of 1.02 per cent and D the average diameter of initial grains.

(3) The driving force of the growth of the recrystallized grain is given by the expression

$$B\sigma(l+c)/Dc$$

at temperatures below the characteristic one and by

$$B\sigma\, l/Dc$$

at the higher than this, where B is a numerical constant with the magnitude of 1.6~1.9, σ the average grain boundary tension. The driving force is less than one hundredth of that expected from the stored energy of the cold worked metal.

(4) With the increase of copper content decreases the characteristic temperature, at which the driving force changes its functional form from $B\sigma(l+c)/Dc$ to $B\sigma l/Dc$.

(5) The relations $\log G_0$ *vs.* $1/T$ for the same driving force as well as for the same samples does not reveal straight lines but curves concave downwards. At temperatures lower than the characteristic the data are approximated by straight lines.

(6) The activation energy determined at rather low temperatures increases, as the addition of small amounts of copper, to a maximum at about 0.1 per cent Cu and then rapidly decreases by the further increase of copper content.

(7) The growth rate is depressed by the dissolutions of precipitates.

(8) The divergences between the existing investigations on the growth rate are reasonably accounted for on the basis of this investigation.

The theoretical consideration of these results based on atomistic model forms another article of paper will be shortly published.

The author wishes to express his thanks to professor S. Takeuchi for his interest and encouragement in this work, and to Mr. S. Ikeda for his helpful assistance throughout the work.